U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE FORM PTO-1390 (Modified) 219902US0XPCT TRANSMITTAL LETTER TO THE UNITED STATES U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR DESIGNATED/ELECTED OFFICE (DO/EO/US) 0708**1**7 CONCERNING A FILING UNDER 35 U.S.C. 371 INTERNATIONAL APPLICATION NO. INTERNATIONAL FILING DATE PRIORITY DATE CLAIMED PCT/EP00/06501 8 July 2000 9 September 1999 (earliest) TITLE OF INVENTION ANTIMICROBIAL ADDITIVES APPLICANT(S) FOR DO/EO/US SOSNA Friedrich et al. Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information: This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include itens (5), (6), (9) and (24) indicated below. 3.  $\boxtimes$  $\boxtimes$ The US has been elected by the expiration of 19 months from the priority date (Article 31). 5.  $\boxtimes$ A copy of the International Application as filed (35 U.S.C. 371 (c) (2)) a. 🗆 is attached hereto (required only if not communicated by the International Bureau). b. 🛛 has been communicated by the International Bureau. ç. 🛘 is not required, as the application was filed in the United States Receiving Office (RO/US). X An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). a. 🛛 is attached hereto. Ъ. 🗆 has been previously submitted under 35 U.S.C. 154(d)(4). 7.  $\boxtimes$ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)) a. 🗀 are attached hereto (required only if not communicated by the International Bureau). b. 🗆 have been communicated by the International Bureau. c. 🗆 have not been made; however, the time limit for making such amendments has NOT expired. have not been made and will not be made. An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)). An English language translation of the annexes to the International Preliminary Examination Report under PCT 10. Article 36 (35 U.S.C. 371 (c)(5)). 11. A copy of the International Preliminary Examination Report (PCT/IPEA/409). 12. A copy of the International Search Report (PCT/ISA/210). Items 13 to 20 below concern document(s) or information included:  $\boxtimes$ An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 13. 14. An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.  $\boxtimes$ 15. A FIRST preliminary amendment. A SECOND or SUBSEQUENT preliminary amendment. 16.  $\times$ 17. A substitute specification.

18. 

A change of power of attorney and/or address letter.

19. A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.

20. A second copy of the published international application under 35 U.S.C. 154(d)(4).

21.  $\square$  A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).

23. Other items or information:

Notice of Priority/PCT/IB/308/Application Data Sheet (2 pages) Form PTO-1449/Letter Regarding Translation of Annexes/Form PTO-1595

	U.S. APPLICATION NO. (IF KNOWN, SEE 37 CF.	R INTERNATIONAL	INTERNATIONAL APPLICATION NO.			ATTORNEY'S DOCKET NUMBER	
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	ASIC NATIONAL FEE ( 37 CFR 1.492 (a) (1) - (5)):				Ī		
	□ Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO						
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i	☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4)						
	☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4)						
	ENTER APPROPRIATE BASIC FEE AMOUNT =					\$890.00	
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	A duplicate copy of this sheet is enclosed.  c.   The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment						
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	d.  Fees are to be charged to a credit card. <b>WARNING:</b> Information on this form may become public. <b>Credit card information should not be included on this form.</b> Provide credit card information and authorization on PTO-2038.						
	NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.						
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### IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF:

FRIEDRICH SOSNA ET AL

: ATTN: APPLICATION DIVISION

SERIAL NO: NEW U.S. PCT APPLN
(Based on PCT NO/EP00/06501)

(Based on PCT NO/EP00/06501)

FILED: HEREWITH

: EXAMINER:

FOR: MICROBICIDAL ADDITIVES

## **PRELIMINARY AMENDMENT**

ASSISTANT COMMISSIONER FOR PATENTS WASHINGTON, D.C. 20231

SIR:

Prior to examination on the merits, please amend the above-identified application as follows.

### IN THE CLAIMS

Please amend the claims as shown on the marked-up copy following this amendment to read as follows:

3. (Amended) The antimicrobial polymer blend as claimed in claim 1, characterized in that the monomer used of the formula I is 2-tert-butylaminoethyl methacrylate, 2-diethylaminoethyl methacrylate, 2-dimethylaminomethyl methacrylate, 2-tert-butylaminoethyl acrylate, 3-dimethylaminopropyl acrylate, 2-diethylaminoethyl acrylate, 2-dimethylaminopropylmethacrylamide, N-3-dimethylaminopropylmethacrylamide, N-3-dimethylaminopropylacrylamide, or N-3-diethylaminopropylacrylamide.

- 4. (Amended) The antimicrobial polymer blend as claimed in claim 1, characterized in that the monomer used of the formula I is 2-tert-butylaminoethyl methacrylate, 2-diethylaminoethyl methacrylate, 2-dimethylaminomethyl methacrylate, 2-tert-butylaminoethyl acrylate, 3-dimethylaminopropyl acrylate, 2-diethylaminoethyl acrylate, 2-dimethylaminopropylmethacrylamide, N-3-dimethylaminopropylmethacrylamide, N-3-dimethylaminopropylacrylamide, or N-3-diethylaminopropylacrylamide.
- 5. (Amended) The antimicrobial polymer blend as claimed in Claim 1, characterized in that the other polymer used comprises polyurethanes, polyolefins, polyethylene, polypropylene, poly-siloxanes, polystyrene, polyacrylates, polymethylmethacrylate, PVC, polyamides or polyterephthalates.
- 6. (Amended) The use of the antimicrobial polymer blends as claimed in Claim 1 for producing items for medical technology.
- 7. (Amended) The use of the antimicrobial polymer blends as claimed in Claim 1 for producing hygiene items.
- 8. (Amended) The use of the antimicrobial polymer blends as claimed in Claim 1 in surface coatings, protective paints, or other coatings.
- 9. (Amended) The use of the antimicrobial polymer blends as claimed in Claim 1 in biocidal formulations.
- 10. (Amended) The use of the antimicrobial polymer blends as claimed in Claim 1 for producing films, tarpaulins, fabrics, or fibers.
- 11. (Amended) The use of the antimicrobial polymer blends as claimed in Claim 1 in formulations for ointments or pastes.
  - 12. (Amended) A process for sterilizing cooling water streams, which comprises

adding antimicrobial polymer blends as claimed in Claim 1 in dispersed form to the cooling water.

### **REMARKS**

Claims 1-12 are active in the present application. Claims 3-12 have been amended to remove multiple dependencies. No new matter is added. An action on the merits and allowance of claims is solicited.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND, MAIER & NEUSTADT, P.C.

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Serial No:
Amendment Filed on:

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### IN THE CLAIMS

- --3. (Amended) The antimicrobial polymer blend as claimed in claim 1[ or 2], characterized in that the monomer used of the formula I is 2-tert-butylaminoethyl methacrylate, 2-diethylaminoethyl methacrylate, 2-dimethylaminomethyl methacrylate, 2-tert-butylaminoethyl acrylate, 3-dimethylaminopropyl acrylate, 2-diethylaminoethyl acrylate, 2-dimethylaminoethyl acrylate, N-3-dimethylaminopropylmethacrylamide, N-3-diethylaminopropylmethacrylamide, or N-3-diethylaminopropylacrylamide.
- 4. (Amended) The antimicrobial polymer blend as claimed in claim 1[ or 2], characterized in that the monomer used of the formula I is 2-tert-butylaminoethyl methacrylate, 2-diethylaminoethyl methacrylate, 2-dimethylaminomethyl methacrylate, 2-tert-butylaminoethyl acrylate, 3-dimethylaminopropyl acrylate, 2-diethylaminoethyl acrylate, 2-dimethylaminopropylmethacrylamide, N-3-dimethylaminopropylmethacrylamide, N-3-dimethylaminopropylacrylamide, or N-3-diethylaminopropylacrylamide.
- 5. (Amended) The antimicrobial polymer blend as claimed in [any of claims 1 to 5] Claim 1, characterized in that the other polymer used comprises polyurethanes, polyolefins,

polyethylene, polypropylene, poly-siloxanes, polystyrene, polyacrylates, polymethylmethacrylate, PVC, polyamides or polyterephthalates.

- 6. (Amended) The use of the antimicrobial polymer blends as claimed in [any of claims 1 to 5] Claim 1 for producing items for medical technology.
- 7. (Amended) The use of the antimicrobial polymer blends as claimed in [any of claims 1 to 5] Claim 1 for producing hygiene items.
- 8. (Amended) The use of the antimicrobial polymer blends as claimed in [any of claims 1 to 5] Claim 1 in surface coatings, protective paints, or other coatings.
- 9. (Amended) The use of the antimicrobial polymer blends as claimed in [any of claims 1 to 5] Claim 1 in biocidal formulations.
- 10. (Amended) The use of the antimicrobial polymer blends as claimed in [any of claims 1 to 5] Claim 1 for producing films, tarpaulins, fabrics, or fibers.
- 11. (Amended) The use of the antimicrobial polymer blends as claimed in [any of claims 1 to 5] Claim 1 in formulations for ointments or pastes.
- 12. (Amended) A process for sterilizing cooling water streams, which comprises adding antimicrobial polymer blends as claimed in [any of claims 1 to 5] Claim 1 in dispersed form to the cooling water.—

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Antimicrobial additives

The invention relates to antimicrobial polymers obtained by polymerizing acryloxyalkylamines. The invention further relates to a process for preparing these antimicrobial polymers and to their use.

It is highly undesirable for bacteria to become established or to spread on the surfaces of piping, or of containers or packaging. Slime layers frequently form and permit sharp rises in microbial populations, and these can lead to persistent impairment of the quality of water, drinks or foods, and even to spoilage of the product and harm to the health of consumers.

Bacteria must be kept away from all fields of life where hygiene is important. This affects textiles for direct body contact, especially in the genital area, and those for the care of the elderly or sick. Bacteria must also be kept away from surfaces of the furniture and equipment used in patient-care areas, especially in areas for intensive care or neonatal care, and in hospitals, especially in the areas where medical intervention takes place, and in isolation wards for critical cases of infection, and also in toilets.

A current method of treating equipment, or the surfaces of furniture or of textiles, to resist bacteria either when this becomes necessary or else as a precautionary measure, is to use chemicals or solutions or mixtures of these which are disinfectants and therefore have fairly broad general antimicrobial action. Chemical agents of this type act nonspecifically and are frequently themselves toxic or irritant, or form degradation products which are hazardous to health. In addition, people frequently exhibit intolerance to these materials once they have become sensitized.

Another method of counteracting surface spread of

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bacteria is to incorporate substances with antimicrobial action into a matrix.

US-A 4 532 269, for example, discloses a terpolymer made from butyl methacrylate, tributyltin methacrylate, and tert-butylaminoethyl methacrylate. This copolymer is used as an antimicrobial paint for ships, and the hydrophilic tert-butylaminoethyl methacrylate present promotes gradual erosion of the polymer and thus releases the highly toxic tributyltin methacrylate, which is the antimicrobial active ingredient.

In these applications, the copolymer prepared with aminomethacrylates is merely a matrix or carrier for added microbicidal ingredients which can diffuse or migrate out of the carrier material. Sooner or later, polymers of this type loose their activity, once the necessary minimum inhibitor concentration (MIC) at the surface has been lost.

US 4 389 502 discloses a polymer composition which comprises polyester, polymethyl methacrylate, cellulose acetate butyrate, and poly(tert-butylaminoethyl methacrylate).

The European patent application 0 862 858 has also disclosed that copolymers of tert-butylaminoethyl methacrylate, a methacrylate with a secondary amino function, has inherent microbicidal properties. Systems developed in the future will again have to be based on novel compositions with improved effectiveness if undesirable resistance phenomena in the microbes are to be avoided, particularly bearing in mind the microbial

The object on which the present invention was based was therefore to develop novel antimicrobial polymers. When used as a coating or covering material, these should prevent bacteria from colonizing surfaces and spreading thereon.

resistance known from antibiotics research.

Surprisingly, it has now been found that

homopolymerization of acryloxyalkylamines or methacryloxyalkylamines gives polymers which are lastingly microbicidal, are not damaged by solvents or physical stresses, and exhibit no migration. There is no need here for the use of other biocidal active ingredients. The surface of the polymers is, of course, for the antimicrobial important action of homopolymers.

The present invention therefore provides antimicrobial polymer blends where one or more antimicrobial polymers which are obtained by polymerizing a monomer of the formula I

$$H_2C$$
 $X-R2-N$ 
 $R3$ 
 $R4$ 

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 $R1 = -H \text{ or } -CH_3$ 

R2 = branched or unbranched aliphatic hydrocarbon radical having from 1 to 5 carbon atoms,

R3 = H, or branched or unbranched aliphatic hydrocarbon radical having from 1 to 7 carbon atoms,

R4 = H, or branched or unbranched aliphatic hydrocarbon radical having from 1 to 7 carbon atoms,

R5 = H, or branched or unbranched aliphatic hydrocarbon radical having from 1 to 7 carbon atoms, and

 $25 \quad X = O, NH, NR5$ 

are mixed with at least one other polymer other than cellulose acetate butyrate and polyester.

Acryloyloxyalkylamines (X = 0) and alkylaminoacryl-30 amides (X = NH) are particularly suitable for preparing the antimicrobial polymers.

The radicals R3 and R4 may be identical or different. If R3 and/or R4 are hydrocarbon groups, these may in particular be methyl, ethyl, isopropyl, n-propyl, or

tert-butyl groups.

Preferred monomers used of the formula I are 2-tertmethacrylate, 2-diethylaminoethyl butylaminoethyl 2-dimethylaminomethyl methacrylate, methacrylate, 2-tert-butylaminoethyl acrylate, 3-dimethylaminopropyl 2-diethylaminoethyl acrylate, acrylate, 2-dimethylacrylate, N-3-dimethylaminopropylmethaminoethyl N-3-diethylaminopropylmethacrylamide, acrylamide, N-3-dimethylaminopropylacrylamide, or N-3-diethylaminopropylacrylamide.

The antimicrobial polymers may be obtained by homopolymerizing monomers of the formula I. The free-radical polymerization advantageously takes place by a chemical route by way of a free-radical initiator, or initiated by radiation. The examples describe typical procedures.

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Examples of blend material, i.e. other polymer with is antimicrobial polymer mixed, the polyurethanes, PVC, polyolefins, such as polyethylene polypropylene, polysiloxanes, polystyrenes, polyacrylates, polymethacrylates, and engineering plastics, e.g. polyamides or polyterephthalates. To obtain adequate antimicrobial action of a polymer blend, the proportion of the antimicrobial polymer of the invention should be from 0.2 to 90% by weight, preferably from 40-90% by weight.

In principle, any of the processes known in the art,

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for example as described in detail by H.-G. Elias, Makromoleküle [Macromolecules], Vol. 2, 5th edition, to prepare et seq., may be used antimicrobial polymer blends. For example, previously formed polymers are mixed in the melt by mixing the pelletized or pulverulent polymers on roll mills, in kneaders, or using extruders. In the case of thermoplastics, this is achieved by heating above the glass transition temperatures or melting points. In the case of solution mixing, the starting materials are separately prepared solutions of the two polymers in the same solvent.

In specific embodiments of the present invention, it is possible for the proportion of the one or more antimicrobial polymers in a blend to be less than 40-90% by weight, e.g. from 0.2 to 70% by weight, preferably from 0.2 to 30% by weight, particularly preferably from 0.2 to 15% by weight, very particularly preferably from 0.2 to 10% by weight.

One preferred process for preparing the antimicrobial polymers and, respectively, polymer blends is freeradical polymerization of monomers of the formula I in solution, using a free-radical initiator. The resultant antimicrobial polymers may, where appropriate after mixing with other polymers, be applied to a surface by known methods, such as dipping, spraying, or spreading. Solvents which have proven successful are ethanol, methanol, water/alcohol mixtures, methyl ethyl ketone, diethvl ether, dioxane, hexane, heptane, toluene, chloroform, dichloromethane, tetrahydrofuran, and acetonitrile, but it is also possible to use other solvents as long as they have sufficient capability for dissolving the polymers and give good wetting of the substrate surfaces. Solutions with polymer contents of from 3 to 20% by weight, for example about 5% by weight, proven successful in practice have

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generally give, in a single pass, coherent coatings which cover the substrate surface and may have a thickness of more than 0.1  $\mu m\,.$ 

- It is also possible for the antimicrobial polymer blends of the invention to be in the form of a melt when applied to the substrates, e.g. by coextrusion, or by way of dipping, spraying, or surface-coating.
- The antimicrobial polymer blends of the invention may moreover also be used as additives or components for formulating polymer blends, inks or paints, surface coatings, or biocides.
- In the case of the polymer blends, a particularly advantageous method is compounding by way of extrusion, where appropriate also by way of coextrusion with other polymers.
- If polymer blends of the invention are used as an additive or component in inks or paints, surface coatings, or biocides, much lower concentrations may be sufficient, e.g. in the range of a few parts per hundred or per thousand.

### Use of the modified polymer substrates

The present invention further provides the use of the antimicrobial polymer blends of the invention producing antimicrobial products, and the resultant The products may comprise products per se. antimicrobial polymers of the invention or consist of these. Products of this type are preferably based on polyurethanes, on polyether polyamides, on on polyesteramides or -imides, on PVC, amides, polyolefins, silicones, on polysiloxanes, on polymethacrylate, or on polyterephthalates, which have surfaces coated with polymers of the invention or have been processed using polymers of the invention, in the

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form of a polymer blend.

Examples of antimicrobial products of this type are machine parts for processing food and drink, components in air conditioning systems, roofing, items for bathroom and toilet use, kitchen items, components for sanitary installations components of animal cages or of animal houses, recreational products for children, components of water systems, packaging for food or drink, operator units (touch panels) of devices, and contact lenses.

The polymer blends of the invention may be used anywhere where importance is placed on surfaces which are as free as possible from bacteria, i.e. are microbicidal, or on surfaces with release properties. Examples of applications of the polymer blends of the invention are in particular surface coatings, protective paints, and other coatings in the following sectors:

- marine: boat hulls, docks, buoys, drilling platforms, ballast water tanks
- construction: roofing, basements, walls, facades,
   greenhouses, sun protection, garden fencing, wood
   protection, tent roof material, fabrics
  - sanitary: public conveniences, bathrooms, shower
    curtains, toilet items, swimming pools, saunas,
    jointing, sealing compounds
- o requisites for daily life: machines, kitchen, kitchen items, sponge pads, recreational products for children, packaging for food or drink, milk processing, drinking water systems, cosmetics
- machine parts: air conditioning systems, ion
   exchangers, process water, solar-powered units, heat
   exchangers, bioreactors, membranes
  - medical technology: contact lenses, diapers, membranes, implants

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- consumer articles: automobile seats, clothing (socks, sports clothing), hospital equipment, door handles, telephone handsets, public conveyances, animal cages, cash registers, carpeting, wallpapers.

The polymer blends may likewise be used as an additive for surface coatings in the maritime sector, in particular for eliminating larval barnacles on boat hulls, and generally as an additive in antifouling paints, particularly in sea water in which salt is present.

The antimicrobial polymer blends of the invention may also be used as additives in formulating cosmetic products, e.g. for pastes or ointments. Here the proportion of polymer blends of the invention may be lowered as far as relatively small numbers of parts per hundred or parts per thousand, depending on the activity of the polymer and the formulation.

The polymer blends of the invention are also used as a biofouling inhibitor in cooling circuits. To prevent damage to cooling circuits by infestation with algae or the circuits would have to be bacteria, frequently or appropriately oversized. In open cooling systems, as are usually found in power plants and in addition of microbicidal chemical plants, the substances such as formalin is not possible. Other microbicidal substances are frequently highly corrosive or form foams, preventing their use in systems of this type.

In contrast, it is possible for polymer blends of the invention to be fed with the other polymers mentioned in finely dispersed form into the process water. The bacteria are killed on contact with the antimicrobial polymers and, if necessary, removed from the system by filtering off the dispersed blend. Deposits of bacteria

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or algae on sections of the plant can thus effectively be prevented. The result of this is a completely novel process for eliminating or reducing biofouling in process water systems.

The present invention therefore also provides a process for sterilizing cooling water streams, by adding antimicrobial polymers or polymer blends of these in dispersed form to the cooling water. For the purposes of the present invention, cooling water includes any process water stream which is used for heating or cooling purposes in closed or open circulating systems.

The dispersed form of the blends may be obtained in the itself, preparation process e.g. by emulsion polymerization, precipitation polymerization, or polymerization, suspension or subsequently by comminuting, e.g. in a jet mill. The size distribution of the resultant particles when they are used is preferably from 0.001 to 3 mm (diameter of particles), firstly providing a large surface for killing the bacteria or algae and secondly enabling, if required, ready separation from the cooling water, filtration. One way of working the process is to remove from the system continuously a proportion (5 to 10%) of the blends used and to replace it with an appropriate amount of fresh material. As an alternative, the number of microbes in the water may be checked, and further copolymer/blend antimicrobial added as Depending on the quality of the water, it is sufficient to use from 0.1 to 100 g of antimicrobial blend per  $m^3$ of cooling water.

The present invention also provides the use for producing hygiene products or items for medical technology, of the polymer substrates modified on the surface using polymer blends of the invention. The statements above concerning preferred materials are

again applicable. Examples of hygiene products of this type are toothbrushes, toilet seats, combs, and packaging materials. For the purposes of the present invention, hygiene items also include articles which can come into contact with many people, for example telephone handsets, stair rails, door handles, window catches, and also grab straps and grab handles in public conveyances. Examples of items for medical technology are catheters, tubing, protective or backing films, and also surgical instruments.

The examples below are given for further description of the present invention, but are not intended to limit the scope of the invention as set out in the claims.

# Example 1:

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60 ml of 2-diethylaminoethyl methacrylate (Aldrich) and 250 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.74 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1.5 l of demineralized water, whereupon the polymeric product After separation of the product precipitates. filtration, the filter residue is washed with 100 ml of a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours.

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### Example 1a:

0.05 g of the product from example 1 is shaken in 20 ml of a test microbial suspension of Staphylococcus

aureus. After a contact time of 15 minutes, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

### Example 1b:

0.05 g of the product from example 1 is shaken in 20 ml of a test microbial suspension of Pseudomonas aeruginosa. After a contact time of 60 minutes, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^3$ .

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#### Example 2:

90 ml of 2-tert-butylaminoethyl methacrylate (Aldrich) and 180 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.745 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1 l of demineralized water, whereupon the polymeric product precipitates. After separation of the product by filtration, the filter residue is washed with 100 ml of a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours.

Example 2a:

0.05 g of the product from example 2 is shaken in 20 ml of a test microbial suspension of Staphylococcus aureus. After a contact time of 15 minutes, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus

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microbes are detectable.

# Example 2b:

0.05 g of the product from example 2 is shaken in 20 ml of a test microbial suspension of Pseudomonas aeruginosa. After a contact time of 60 minutes, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^2$ .

## Example 3:

20 ml of N-3-dimethylaminopropylacrylamide (Aldrich) and 70 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.2 g of azobisisobutyronitrile dissolved in 5 ml of ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 0.5 l of demineralized water, whereupon the polymeric product precipitates. After separation of the product by filtration, the filter residue is washed with 100 ml of a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours.

### Example 3a:

0.05 g of the product from example 3 is shaken in 20 ml of a test microbial suspension of Staphylococcus aureus. After a contact time of 15 minutes, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^3$ .

# Example 3b:

0.05 g of the product from example 3 is shaken in 20 ml

of a test microbial suspension of Pseudomonas aeruginosa. After a contact time of 60 minutes, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^3$ .

### Example 4:

10 g of the polymer from example 1 are heated to 165°C. This heated polymer is then mixed with 3 g of polymethyl methacrylate (Aldrich) which had likewise been heated in advance to 165°C. The two polymers are very thoroughly mixed and cooled to room temperature at a rate of 20°C per hour.

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### Example 4a:

0.05 g of the product from example 4 is shaken in 20 ml of a test microbial suspension of Staphylococcus aureus. After a contact time of 15 minutes, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^3$ .

### 25 Example 4b:

0.05 g of the product from example 4 is shaken in 20 ml of a test microbial suspension of Pseudomonas aeruginosa. After a contact time of 60 minutes, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^4$ .

### Example 5:

90 ml of 2-tert-butylaminoethyl methacrylate (Aldrich) and 180 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.745 g of azobisisobutyronitrile dissolved in 20 ml of

ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1 l of demineralized water, whereupon the polymeric product precipitates. After separation of the product filtration, the filter residue is washed with 100 ml of a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 5 q of the product are dissolved in 32 g of diisononyl phthalate. 64 g of polyvinyl chloride pellets are then added to this mixture, and the mixture is intimately mixed until it becomes a paste. 20 g of the resultant paste are applied to a metal sheet, using a doctor, in such a way as to give a layer of 0.7 mm thickness. The sheet covered by the paste is then heated to 200°C for 2 minutes, whereupon the paste gels, giving plasticized PVC film.

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### Example 5a:

A piece of the plasticized PVC film from example 5, dimensions  $3 \times 3$  cm, is placed on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

#### Example 5b:

A piece of the plasticized PVC film from example 5, dimensions  $3 \times 3$  cm, is placed on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 4 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined.

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After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

# Example 6:

90 ml of 2-tert-butylaminoethyl methacrylate (Aldrich) and 180 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.745 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1 l of demineralized water, whereupon the polymeric product After separation of the product precipitates. filtration, the filter residue is washed with 100 ml of a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 2 g of the product are dissolved in 32 g of diisononyl phthalate. 64 g of polyvinyl chloride pellets are then added to this mixture, and the mixture is intimately mixed until it becomes a paste. 20 g of the resultant paste are applied to a metal sheet, using a doctor, in such a way as to give a layer of 0.7 mm thickness. The sheet covered by the paste is then heated to 200°C for giving paste gels, whereupon the 2 minutes, plasticized PVC film.

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### Example 6a:

A piece of the plasticized PVC film from example 6, dimensions  $3 \times 3$  cm, is placed on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

### Example 6b:

A piece of the plasticized PVC film from example 6, dimensions  $3 \times 3$  cm, is placed on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 4 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^3$ .

#### Example 7:

90 ml of 2-tert-butylaminoethyl methacrylate (Aldrich) and 180 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.745 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1 l of demineralized water, whereupon the polymeric product precipitates. After separation of the product by filtration, the filter residue is washed with 100 ml of a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 5 g of the product are stirred into 95 g of Rowacryl G-31293

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acrylic surface coating from the company ROWA.

## Example 7a:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the acrylic surface coating from example 7 and then dried for 24 hours at  $35^{\circ}$ C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

## Example 7b:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the acrylic surface coating from example 7 and then dried for 24 hours at 35°C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After this time, remaining Pseudomonas no aeruginosa microbes are detectable.

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#### Example 8:

90 ml of 2-tert-butylaminoethyl methacrylate (Aldrich) and 180 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.745 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this

time, the reaction mixture is stirred into 1 l of demineralized water, whereupon the polymeric product precipitates. After separation of the product by filtration, the filter residue is washed with 100 ml of a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 2 g of the product are stirred into 98 g of Rowacryl G-31293 acrylic surface coating from the company ROWA.

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### Example 8a:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the acrylic surface coating from example 8 and then dried for 24 hours at  $35^{\circ}$ C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

#### 25 Example 8b:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the acrylic surface coating from example 8 and then dried for 24 hours at 35°C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^3$ .

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### Example 9:

90 ml of 2-tert-butylaminoethyl methacrylate (Aldrich) and 180 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.745 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this the reaction mixture is stirred into 1 l of demineralized water, whereupon the polymeric product After separation of the product. precipitates. filtration, the filter residue is washed with 100 ml of a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 5 g of the product are stirred into 95 g of Plextol D 510 from the aqueous dispersion company PolymerLatex, an methacrylate-acrylate copolymer.

### 20 Example 9a:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the dispersion from example 9 and then dried for 24 hours at 35°C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

#### Example 9b:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the dispersion from example 9 and then dried for 24 hours at  $35^{\circ}$ C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a

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test microbial suspension of Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^2$ .

### Example 10:

90 ml of 2-tert-butylaminoethyl methacrylate (Aldrich) and 180 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.745 q of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1 l of demineralized water, whereupon the polymeric product separation of the product precipitates. After filtration, the filter residue is washed with 100 ml of a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 2 g of the product are stirred into 98 g of Plextol D 510 from the PolymerLatex, an aqueous dispersion methacrylate-acrylate copolymer.

#### Example 10a:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the dispersion from example 10 and then dried for 24 hours at  $35^{\circ}$ C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

### Example 10b:

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Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the dispersion from example 10 and then dried for 24 hours at  $35^{\circ}$ C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^2$ .

### 15 Example 11:

90 ml of 2-tert-butylaminoethyl methacrylate (Aldrich) and 180 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.745 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1 l of demineralized water, whereupon the polymeric product 25 precipitates. After separation of the product by filtration, the filter residue is washed with 100 ml of a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 1 g of the product is dissolved in 99 g of ethanol. Six cotton 30 pads, each of diameter 3 cm, are immersed for 1 second into this solution, removēd, and dried at temperature for 24 hours.

# 35 Example 11a:

Cotton pads from example 11 are inoculated with, respectively, Chlorella sp., Trentepohlia sp., Gloeocapsa sp., Calothrix sp., and Aspergillus niger.

These specimens are then placed in an incubator for 3 weeks. Unlike control specimens run simultaneously, no growth is detectable on any of the coated absorbent cotton pads.

Example 12:

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60 ml of 2-diethylaminoethyl methacrylate (Aldrich) and 250 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.74 g of azobisisobutyronitrile dissolved in of 20 ml methyl ketone is then slowly added dropwise, stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1.5 l of demineralized water, whereupon the polymeric product After separation of the product precipitates. filtration, the filter residue is washed with 100 ml of a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 2 g of the product are dissolved in 10 g of tetrahydrofuran and applied, using a 100 micrometer doctor, to an aluminum sheet of 0.5 cm thickness and dimensions  $2 \times 2$  cm. The sheet is then dried for 24 hours at 50°C.

Example 12a:

The aluminum sheet from example 12 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

### Example 12b:

The aluminum sheet from example 12 is placed, with its

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coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and subjected to shaking. After a contact time of 4 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Pseudomonas aeruginosa microbes are detectable.

## 10 Example 13:

90 ml of 2-tert-butylaminoethyl methacrylate (Aldrich) and 180 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.745 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1 l of demineralized water, whereupon the polymeric product After separation of the product precipitates. filtration, the filter residue is washed with 100 ml of a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 2 g of the product are dissolved in 10 g of tetrahydrofuran and applied, using a 100 micrometer doctor, to an aluminum sheet of  $0.5~\mathrm{cm}$  thickness and dimensions  $2\times2~\mathrm{cm}$ . The sheet is then dried for 24 hours at 50°C.

# 30 Example 13a:

The aluminum sheet from example 13 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are

detectable.

### Example 13b:

The aluminum sheet from example 13 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and subjected to shaking. After a contact time of 4 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Pseudomonas aeruginosa microbes are detectable.

### Example 14:

15 20 ml of 3-dimethylaminopropyl acrylate (Aldrich) and 70 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.2 g of azobisisobutyronitrile dissolved in 5 ml of methyl ketone is then slowly added dropwise, stirring. The mixture is heated to 70°C and stirred at 20 this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 0.5 l of demineralized water, whereupon the polymeric product precipitates. After separation of the product filtration, the filter residue is washed with 100 ml of 25. a 10% strength solution of ethanol in water in order to remove any residual monomers still present. The product is then dried in vacuo at  $50\,^{\circ}\text{C}$  for 24 hours. 2 g of the product are dissolved in 10 g of tetrahydrofuran and 30 applied, using a 100 micrometer doctor, to an aluminum sheet of 0.5 cm thickness and dimensions  $2 \times 2$  cm. The sheet is then dried for 24 hours at 50°C.

### Example 14a:

The aluminum sheet from example 14 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and subjected to shaking. After

a contact time of 4 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

### Example 14b:

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The aluminum sheet from example 14 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and subjected to shaking. After a contact time of 8 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Pseudomonas aeruginosa microbes are detectable.

### Example 15:

10 g of the polymer from example 1 are heated to  $165^{\circ}$ C. This heated polymer is then mixed with 3 g of polymethyl methacrylate (Aldrich) which had likewise been heated in advance to  $165^{\circ}$ C. The two polymers are very thoroughly mixed, applied to an aluminum sheet with a thickness of 0.5 cm and dimensions 2 x 2 cm, and cooled to room temperature at a rate of 20°C per hour.

### Example 15a:

The aluminum sheet from example 15 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and subjected to shaking. After a contact time of 4 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

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### Example 15b:

The aluminum sheet from example 15 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and subjected to shaking. After a contact time of 8 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Pseudomonas aeruginosa microbes are detectable.

### Example 16:

of dimethylaminopropylmethacrylamide (Aldrich) and 250 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.6 g 15 of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1.5 l of 20 cyclohexane, whereupon the polymeric product precipitates. After separation of the product filtration, the filter residue is washed with 100 ml of n-hexane in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 2 g of the product are dissolved in 10 g of tetrahydrofuran and applied, using a 100 micrometer doctor, to an aluminum sheet of 0.5 cm thickness and dimensions  $2 \times 2$  cm. The sheet is then dried for 24 hours at 50°C.

#### Example 16a:

The aluminum sheet from example 16 is placed, with its coated side upward, on the base of a glass beaker which 20 ml of a test microbial suspension of Staphylococcus aureus, and subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in

the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

# 5 Example 16b:

The aluminum sheet from example 16 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and subjected to shaking. After a contact time of 4 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from 10<sup>7</sup> to 10<sup>3</sup> microbes per ml.

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#### Example 17:

50 ml of diethylaminopropylmethacrylamide (Aldrich) and 250 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.6 g of azobisisobutyronitrile dissolved in 20 ml of methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1.5 l of cyclohexane, whereupon the polymeric precipitates. After separation of the product filtration, the filter residue is washed with 100 ml of n-hexane in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 2 g of the product are dissolved in 10 g of tetrahydrofuran and applied, using a 100 micrometer doctor, to an aluminum sheet of 0.5 cm thickness and dimensions  $2 \times 2$  cm. The sheet is then dried for 24 hours at 50°C.

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### Example 17a:

The aluminum sheet from example 17 is placed, with its coated side upward, on the base of a glass beaker which

contains 20 ml of a test microbial suspension of Staphylococcus aureus, and subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

### Example 17b:

The aluminum sheet from example 17 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and subjected to shaking. After a contact time of 4 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from 10<sup>7</sup> to 10<sup>3</sup> microbes per ml.

#### 20 Example 18:

N-3-dimethylaminopropylacrylamide (Aldrich) and 250 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.6 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, 25 stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1.5 l of cyclohexane, whereupon the polymeric product After 30 precipitates. separation of the product by filtration, the filter residue is washed with 100 ml of n-hexane in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 2 g of the product are dissolved in 10 g of tetrahydrofuran and applied, using a 100 micrometer 35 doctor, to an aluminum sheet of 0.5 cm thickness and dimensions  $2 \times 2$  cm. The sheet is then dried 24 hours at 50°C.

### Example 18a:

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The aluminum sheet from example 18 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and subjected to shaking. After a contact time of 4 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

### Example 18b:

The aluminum sheet from example 18 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and subjected to shaking. After a contact time of 8 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from 10<sup>7</sup> to 10<sup>4</sup> microbes per ml.

#### Example 19:

10 g of the polymer from example 16 are heated to 165°C. This heated polymer is then mixed with 3 g of polymethyl methacrylate (Aldrich) which had likewise been heated in advance to 165°C. The two polymers are very thoroughly mixed, applied to an aluminum sheet with a thickness of 0.5 cm and dimensions 2 x 2 cm, and cooled to room temperature at a rate of 20°C per hour.

#### Example 19a:

The aluminum sheet from example 19 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and subjected to shaking. After a contact time of 4 hours, 1 ml of the test microbial

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suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

Example 19b:

The aluminum sheet from example 19 is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and subjected to shaking. After a contact time of 8 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from 10<sup>7</sup> to 10<sup>4</sup> microbes per ml.

## Example 20:

50 ml of dimethylaminopropylmethacrylamide (Aldrich) and 250 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.6 g 20 of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1.51 of 25 whereupon cyclohexane, the polymeric precipitates. After separation of the product by filtration, the filter residue is washed with 100 ml of n-hexane in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 30 24 hours. 6 g of the product are dissolved in 32 g of diisononyl phthalate. 64 g of polyvinyl chloride pellets are then added to this mixture, and the mixture is intimately mixed until it becomes a paste. 20 q of the resultant paste are applied to a metal sheet, using 35 a doctor, in such a way as to give a layer of 0.7 mm thickness. The sheet covered by the paste is then heated to 200°C for 2 minutes, whereupon the paste

gels, giving a plasticized PVC film.

### Example 20a:

A piece of the plasticized PVC film from example 20, dimensions 3 x 3 cm, is placed on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

## Example 20b:

15 A piece of the plasticized PVC film from example 20, dimensions  $3 \times 3$  cm, is placed on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 4 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^4$  microbes per ml.

#### 25 **Example 21:**

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dimethylaminopropylmethacrylamide (Aldrich) and 250 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.6 q of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1.5 l of cyclohexane, whereupon the polymeric product separation of the product by precipitates. After filtration, the filter residue is washed with 100 ml of n-hexane in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for

24 hours. 2 g of the product are dissolved in 32 g of diisononyl phthalate. 64 g of polyvinyl chloride pellets are then added to this mixture, and the mixture is intimately mixed until it becomes a paste. 20 g of the resultant paste are applied to a metal sheet, using a doctor, in such a way as to give a layer of 0.7 mm thickness. The sheet covered by the paste is then heated to 200°C for 2 minutes, whereupon the paste gels, giving a plasticized PVC film.

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#### Example 21a:

A piece of the plasticized PVC film from example 21, dimensions  $3 \times 3$  cm, is placed on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

#### Example 21b:

A piece of the plasticized PVC film from example 21, dimensions  $3 \times 3$  cm, is placed on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 4 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^3$ .

#### Example 22:

50 ml of diethylaminopropylmethacrylamide (Aldrich) and 250 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.6 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, with

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stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1.5 l of cyclohexane, whereupon the polymeric product precipitates. After separation of the product filtration, the filter residue is washed with 100 ml of n-hexane in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 2 g of the product are dissolved in 32 g of diisononyl phthalate. 64 g of polyvinyl chloride pellets are then added to this mixture, and the mixture is intimately mixed until it becomes a paste. 20 g of the resultant paste are applied to a metal sheet, using a doctor, in such a way as to give a layer of 0.7 mm thickness. The sheet covered by the paste is then heated to 200°C for 2 minutes, whereupon the paste gels, giving a plasticized PVC film.

## Example 22a:

A piece of the plasticized PVC film from example 22, dimensions 3 x 3 cm, is placed on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

## 30 Example 22b:

A piece of the plasticized PVC film from example 22, dimensions 3 × 3 cm, is placed on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 4 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has

fallen from  $10^7$  to  $10^3$ .

### Example 23:

50 ml of dimethylaminopropylmethacrylamide (Aldrich) and 250 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.6 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at 10 this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1.5 l of cyclohexane, whereupon the polymeric product precipitates. After separation of the product by filtration, the filter residue is washed with 100 ml of n-hexane in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 5 g of the product are stirred into 95 g of Rowacryl G-31293 acrylic surface coating from company ROWA.

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## Example 23a:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the acrylic surface coating from example 23 and then dried for 24 hours at 35°C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

#### 35 Example 23b:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the acrylic surface coating from example 23 and then dried for 24 hours at 35°C in a

drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which 20 ml of a test microbial suspension of contains Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^4$ .

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### Example 24:

of N-3-dimethylaminopropylacrylamide (Aldrich) and 250 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.6 g of azobisisobutyronitrile dissolved in 20 ml of ethyl 15. methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1.5 l of cyclohexane, whereupon the polymeric 20 precipitates. After separation of the product filtration, the filter residue is washed with 100 ml of n-hexane in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 2 g of the product are stirred into 98 g of Rowacryl G-31293 acrylic surface coating from company ROWA.

#### Example 24a:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm 30 is painted with the acrylic surface coating from example 24 and then dried for 24 hours at 35°C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which 20 ml of a test microbial suspension 35 Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of

microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

#### 5 Example 24b:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the acrylic surface coating from example 24 and then dried for 24 hours at 35°C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^3$ .

#### Example 25:

of dimethylaminopropylmethacrylamide (Aldrich) 20 and 250 ml of ethanol are charged to a three-necked flask and heated to 65°C under a stream of argon. 0.6 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, with stirring. The mixture is heated to 70°C and stirred at 25 this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1.5 l of cyclohexane, whereupon the polymeric product After separation of the product precipitates. 30 filtration, the filter residue is washed with 100 ml of n-hexane in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for 24 hours. 5 g of the product are stirred into 95 g of Plextol D 510 from the company PolymerLatex, an aqueous dispersion of a methacrylate-acrylate copolymer.

#### Example 25a:

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Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm

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is painted with the dispersion from example 25 and then dried for 24 hours at 35°C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

## Example 25b:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the dispersion from example 25 and then dried for 24 hours at  $35^{\circ}$ C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^4$ .

## 25 Example 26:

N-3-dimethylaminopropylacrylamide and 250 ml of ethanol are charged to a three-necked flask and heated to  $65^{\circ}\text{C}$  under a stream of argon. 0.6 g of azobisisobutyronitrile dissolved in 20 ml of ethyl methyl ketone is then slowly added dropwise, 30 stirring. The mixture is heated to 70°C and stirred at this temperature for 72 hours. After expiry of this time, the reaction mixture is stirred into 1.5 l of cyclohexane, whereupon the polymeric 35 precipitates. After separation of the product filtration, the filter residue is washed with 100 ml of n-hexane in order to remove any residual monomers still present. The product is then dried in vacuo at 50°C for

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24 hours. 2 g of the product are stirred into 98 g of Plextol D 510 from the company PolymerLatex, an aqueous dispersion of a methacrylate-acrylate copolymer.

## 5 Example 26a:

Using a brush, an aluminum sheet of dimensions 5 x 5 cm is painted with the dispersion from example 26 and then dried for 24 hours at 35°C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Staphylococcus aureus, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, no remaining Staphylococcus aureus microbes are detectable.

## Example 26b:

Using a brush, an aluminum sheet of dimensions  $5 \times 5$  cm is painted with the dispersion from example 26 and then dried for 24 hours at 35°C in a drying cabinet. This aluminum sheet is placed, with its coated side upward, on the base of a glass beaker which contains 20 ml of a test microbial suspension of Pseudomonas aeruginosa, and is subjected to shaking. After a contact time of 2 hours, 1 ml of the test microbial suspension is removed, and the number of microbes in the test mixture is determined. After expiry of this time, the number of microbes has fallen from  $10^7$  to  $10^3$ .

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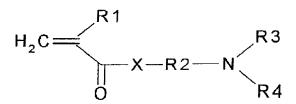
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## What is claimed is:

 An antimicrobial polymer blend, characterized in that

one or more antimicrobial polymers each obtainable by polymerizing a monomer of the formula I



where

10 R1 = -H or  $-CH_3$ 

R2 = branched or unbranched aliphatic hydrocarbon radical having from 1 to 5 carbon atoms,

R3 = H, or branched or unbranched aliphatic hydrocarbon radical having from 1 to 7 carbon atoms,

R4 = H, or branched or unbranched aliphatic hydrocarbon radical having from 1 to 7 carbon atoms,

R5 = H, or branched or unbranched aliphatic hydrocarbon radical having from 1 to 7 carbon atoms, and

X = O, NH, NR5

are mixed with at least one other polymer other than cellulose acetate butyrate and polyesters.

- The antimicrobial polymer blend as claimed in claim 1, characterized in that the polymer blend is composed of from 0.2 to 90% by weight of one or more antimicrobial polymers.
- 3. The antimicrobial polymer blend as claimed in claim 1 or 2, characterized in that

the monomer used of the formula I is 2-tert-butylaminoethyl methacrylate, 2-diethylaminoethyl methacrylate, 2-dimethylaminomethyl methacrylate, 2-tert-butylaminoethyl acrylate, 3-dimethylamino-propyl acrylate, 2-diethylaminoethyl acrylate, 2-dimethylaminoethyl acrylate, N-3-dimethylamino-propylmethacrylamide, N-3-diethylaminopropylmethacrylamide, N-3-diethylaminopropylacrylamide, or N-3-diethylaminopropylacrylamide.

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- 4. The antimicrobial polymer blend as claimed in claim 1 or 2, characterized in that the monomer used of the formula I is 2-tert-butylaminoethyl methacrylate, 2-diethylaminoethyl methacrylate, 2-dimethylaminomethyl methacrylate, 2-tert-butylaminoethyl acrylate, 3-dimethylaminopropyl acrylate, 2-diethylaminoethyl acrylate, 2-dimethylaminoethyl acrylate, N-3-dimethylaminopropylmeth-acrylamide, N-3-dimethylaminopropylmeth-acrylamide, N-3-dimethylaminopropylacrylamide, or
- 5. The antimicrobial polymer blend as claimed in any of claims 1 to 4, characterized in that the other polymer used comprises polyurethanes, polyolefins, polyethylene, polypropylene, polysiloxanes, polystyrene, polyacrylates, polymethyl methacrylate, PVC, polyamides or polyterephthalates.

N-3-diethylaminopropylacrylamide.

- 6. The use of the antimicrobial polymer blends as claimed in any of claims 1 to 5 for producing items for medical technology.
  - 7. The use of the antimicrobial polymer blends as claimed in any of claims 1 to 5 for producing hygiene items.

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- 8. The use of the antimicrobial polymer blends as claimed in any of claims 1 to 5 in surface coatings, protective paints, or other coatings.
- 5 9. The use of the antimicrobial polymer blends as claimed in any of claims 1 to 5 in biocidal formulations.
- 10. The use of the antimicrobial polymer blends as claimed in any of claims 1 to 5 for producing films, tarpaulins, fabrics, or fibers.
  - 11. The use of the antimicrobial polymer blends as claimed in any of claims 1 to 5 in formulations for ointments or pastes.
  - 12. A process for sterilizing cooling water streams, which comprises adding antimicrobial polymer blends as claimed in any of claims 1 to 5 in dispersed form to the cooling water.



(43) Internationales Veröffentlichungsdatum 15. März 2001 (15.03.2001)

PCT

## (10) Internationale Veröffentlichungsnummer WO 01/18077 A1

(51) Internationale Patentklassifikation7: 20/60, A01N 33/12

C08F 20/34.

(21) Internationales Aktenzeichen:

PCT/EP00/06501

(22) Internationales Anmeldedatum:

8. Juli 2000 (08.07.2000)

(25) Einreichungssprache:

Deutsch

(26) Veröffentlichungssprache:

Deutsch

(30) Angaben zur Priorität:

100 22 453.9

DE

199 43 182.5

9. September 1999 (09.09.1999) 9. Mai 2000 (09.05.2000)

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(84) Bestimmungsstaaten (regional): europäisches Patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

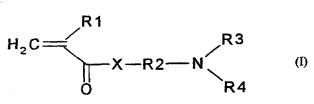
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Mit internationalem Recherchenbericht.

Zur Erklärung der Zweibuchstaben-Codes, und der anderen Abkürzungen wird auf die Erklärungen ("Guidance Notes on Codes and Abbreviations") am Anfang jeder regulären Ausgabe der PCT-Gazette verwiesen.

(54) Title: MICROBICIDAL ADDITIVES

(54) Bezeichnung: ANTIMIKROBIELLE ZUSATZSTOFFE



(57) Abstract: The invention relates to microbicidal polymers and polymer blends, obtained by the polymerization of a monomer of formula (I), wherein R1 = -H or -CH<sub>3</sub>, R2 = a branched or unbranched aliphatic hydrocarbon radical with between 1 and 5 carbon atoms, R3 = H, a branched or unbranched aliphatic hydrocarbon radical with between 1 and 7 carbon atoms and R4 = H, a branched or unbranched aliphatic hydrocarbon radical with between 1 and 7 carbon

atoms, R5 = H, a branched or unbranched aliphatic hydrocarbon radical with between 1 and 7 carbon atoms, X = O, NH, NR5 and by the optional subsequent mixing of said polymers with at least one additional polymer. The microbicidal polymers or polymer blends can be used e.g. as a coating for articles of hygiene or medical articles, or used in lacquers or protective coatings. They can also be used in a method for preventing/reducing biological fouling in water systems.

(57) Zusammenfassung: Die Erfindung betrifft antimikrobielle Polymere und Polymerblends, die durch Polymerisation eines Monomeren der Formel (I) mit R1 = -H oder -CH3, R2 = verzweigter oder unverzweigter aliphatischer Kohlenwasserstoffrest mit 1 bis 5 Kohlenstoffatomen, R3 = H, verzweigter oder unverzweigter aliphatischer Kohlenwasserstoffrest mit 1 bis 7 Kohlenstoffatomen und R4 = H, verzweigter oder unverzweigter aliphatischer Kohlenwasserstoffrest mit 1 bis 7 Kohlenstoffatomen, R5 == H, verzweigter oder unverzweigter aliphatischer Kohlenwasserstoffrest mit 1 bis 7 Kohlenstoffatomen, X = O, NH, NR5 und ggf. nachfolgende Vermischung mit mindestens einem weiteren Polymeren hergestellt werden. Die antimikrobiellen Polymere oder Blends können zur Herstellung von Hygieneartikeln oder medizintechnischen Artikeln, z.B.als Beschichtung sowie in Lacken oder Schutzanstrichen verwendet werden. Des weiteren können sie in einem Verfahren zur Vermeidung/Verringerung von Biofouling in Wassersystemen eingesetzt werden.

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# Declaration and Power of Attorney for Patent Application Erklärung für Patentanmeldungen mit Vollmacht

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(Number) (Nummer)	(Country) (Land)		(Day/Month/Year Filed) (Tag/Monat/Jahr der Anmeldung)	Yes Ja	No Nein
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German Language Declaration

	<del></del>
Vor- und Zuname des dritten Miterfinders (falls Zutreffend)	Full name of third joint inventor, if any
	Beate KOSSMANN
Unterschrift des dritten Erfinders Datum	Third inventor's signature Date
<b>\</b>	7054 Down 11. FEB. 2502
Wohnsitz	Residence
2(1)	Hagen
Staatsangehörigkeit	Citizenship
Staatsangenongken	1 1 2 - 1//
	German
Postanschrift	Post Office Address
	Ribbertstrasse 13, 58091 Hagen, GERMANY
Vor- und Zuname des vierten Miterfinders (falls Zutreffend)	Full name of fourth joint inventor, if any
Unterschrift des vierten Erfinders Datum	Fourth inventor's signature Date
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M-L1-	Desidence
Wohnsitz	Residence
Staatsangehörigkeit	Citizenship
·	
Postanschrift	Post Office Address
Vor- und Zuname des fünften Miterfinders (falls Zutreffend)	Full name of fifth joint inventor, if any
VOI- UND ZUNAME DES JUNIEN MIREMINDES (TAIIS ZUNEMERA)	Pull flame of filtri joint inventor, if any
The state of the first of the state of the s	
Unterschrift des fünften Erfinders Datum	Fifth inventor's signature Date
Wohnsitz	Residence
·	
Staatsangehörigkeit	Citizenship
Postanschrift	Post Office Address
Vor- und Zuname des sechsten Miterfinders (falls Zutreffend)	From the second at the trade to the trade of the second at
vor- und zuname des sechsten wilterlinders (falls zutretiend)	Full name of sixth joint inventor, if any
Unterschrift des sechsten Erfinders Datum	Sixth inventor's signature Date
·	
Wohnsitz	Residence
Staatsangehörigkeit	Citizenship
· · · · · · · · · · · · · · · · · · ·	Old Street, St
Postanschrift	Post Office Address
FOStalischin	Post Office Address

(Im Falle-dritter und weiterer Miterfinder sind die entsprechenden Informationen und Unterschriften hinzuzufugen.)

(Supply similar information and signature for third and subsequent joint inventors.)